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STUDY OF MARITIME AEROSOLS. REPORT TO ENVIRONMENTAL PROTECTION AGENCY. DIVISION OF METEOROLOGY, RESEARCH TRIANGLE PARK, NORTH CAROLINA

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16. Abstracts

The results are presented of a research project which was conducted for the purpose of surveying the atmospheric aerosol concentration over the seas, using the information gathered to evaluate the sources and sinks of these particles, and of beginning a chronology of such observations. A portable, photoelectric aerosol detector was supplied to the navigation officer of each cooperating ship. An aerosol observation was made four times each day. These observations were logged into the "additional message groups" column of a weather data form and mailed to the investigators on return to port. This data was then logged in as a function of position and date. Additional data inputs were received from oceanographic ships, ocean station, and island observers. The aerosol concentration position, time, and meteorological data were key punched. After initial review and analysis of the data by the investigators, computer analysis can be performed to attempt to relate variations in aerosol concentration to meteorological parameters.

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STUDY OF MARITIME AEROSOLS

REPORT TO ENVIRONMENTAL PROTECTION AGENCY

DIVISION OF METEOROLOGY

RESEARCH TRIANGLE PARK, NORTH CAROLINA

CONTRACT 70-64

1 SEPTEMBER 1972

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II

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Introduction:

The purpose of this research is to survey the atmospheric aerosol concentration over the seas, use the information gathered to evaluate the sources and sinks of these particles, and to begin a chronology of such observations. The program was initiated as an inexpensive alternative to the oceanographic cruise as a method for gathering atmospheric data over the seas. It is a joint venture of the State University Maritime College and the Atmospheric Sciences Research Center, drawing on the expertise of ASRC to provide necessary instrumentation, and the facilities and graduate contacts of the Maritime College for logistic support.

A portable, photo electric aerosol detector is supplied to the navigation officer of each cooperating ship. An aerosol observation is made four times per day, in conjunction with the NOAA synoptic weather observation, by the watch officer on the bridge. The observations are logged in the "additional message groups" column of a carbon copy of the weather data form, and the form mailed to the investigators on return to port. This data is then logged as a function of position and date in a set of log books. Additional data inputs are received from oceanographic ships, ocean station, and island observers.

The aerosol concentration, position, time, and meteorological data are then key punched at the SUMC Computer Center. After initial review and analysis of the data by the investigators,

computer analysis can be performed to attempt to relate variations in aerosol concentration to meteorological parameters. The "data bank" accumulated in the past two years is quite sizeable, and permits several analysis, as reported in subsequent sections.

The observations made so far have only measured the number concentration of aerosol particles, in the interest of simplicity and reliability. This simplicity has allowed us to obtain thin coverage of a very large area, over a very short period, to delineate the best areas for intensive study with more sophisticated equipment, and to begin to have an understanding of some of the meteorological sinks for aerosol particles.

The data obtained as a result of our first year's experience, and reported in our first report, "Comprehensive Study of Maritime Aerosols," (Hogan and Degani, submitted 5 August 1971) allowed preparation of a map of aerosol distributions over the North Atlantic, and provided some insight to the activity of storm systems as aerosol sinks. The data acquired since that time has allowed refinement of the original North Atlantic map, preparation of map showing Pacific distributions, and by reference to earlier work in literature, has allowed the beginning of an aerosol chronology of the Atlantic area.

COVERAGE OF OBSERVER NETWORK

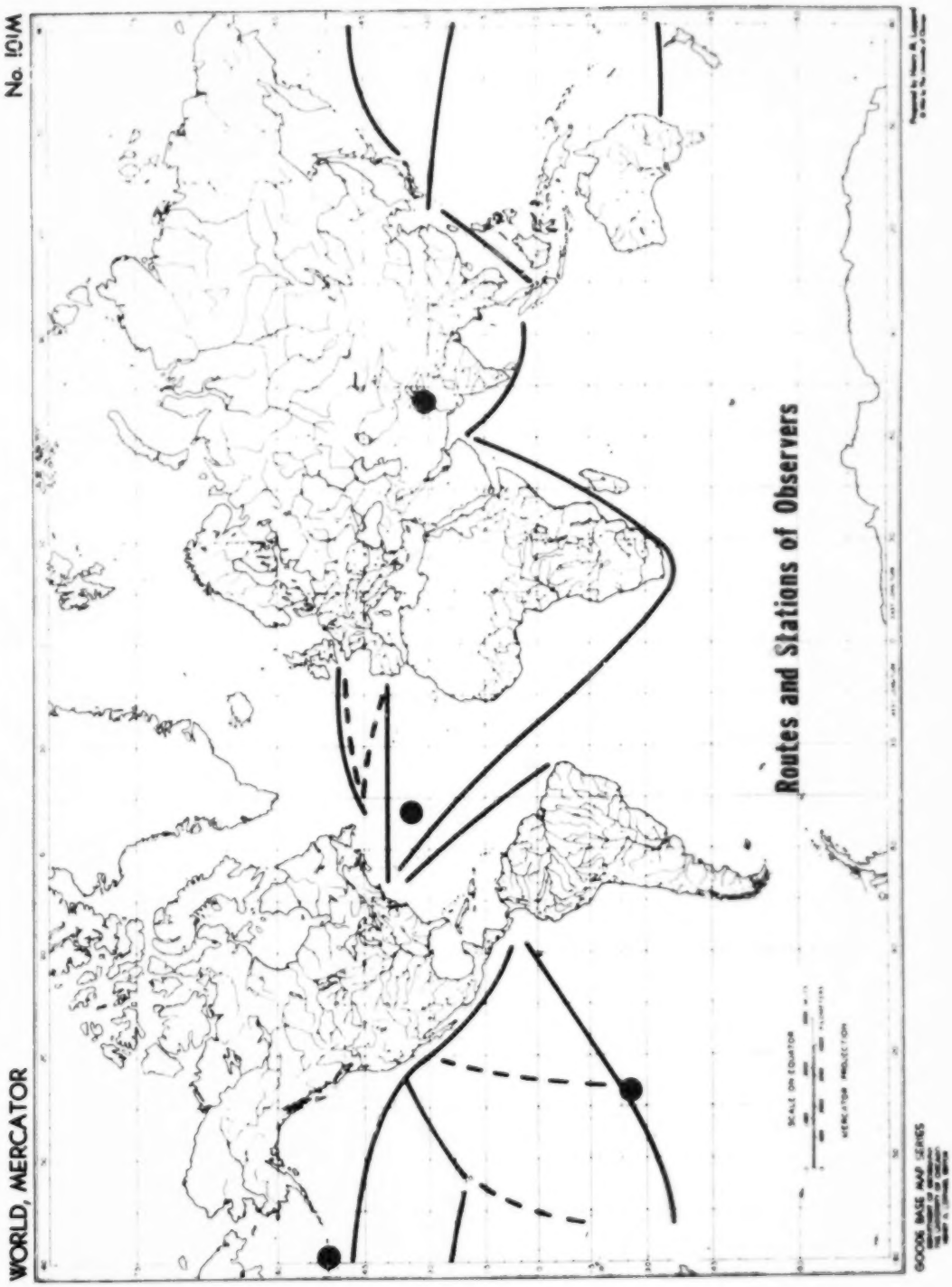
The routes traveled by cooperating observers are shown as Figure I, with merchant trade routes shown as heavy lines, and routes of oceanographic ships as broken lines. Additional fixed point data has been obtained from Ocean Station Echo, Pitcairn Island and Shemya Island.

The mid-latitude coverage obtained in this cooperative program is fairly good. The addition of oceanographic ships to the observation network has given coverage to two areas not routinely covered in the Pacific, and has provided coverage parallel to the trade routes in the North Atlantic.

Several areas have had no coverage during the experiment period. These include the entire region north of 50°N latitude, the southern hemisphere south of 40°S latitude, and almost the entire mid Indian Ocean. Any subsequent program should stress observations in these areas which may be quite important aerosol sinks.

Coverage has been frequent (monthly) on the North Atlantic, and bi-monthly to quarterly on other routes. This frequency has provided sufficient data to prepare a general distribution of aerosol concentrations over the seas, but is not sufficiently frequent to determine seasonal changes. Additional data from island and ocean stations may also be of assistance in determining seasonal changes.

FIGURE I


No. 101M
WORLD, MERCATOR
Routes and Stations of Observers
SCALE ON EQUATOR
MERCATOR PROJECTION
GOODE BASE MAP SERIES

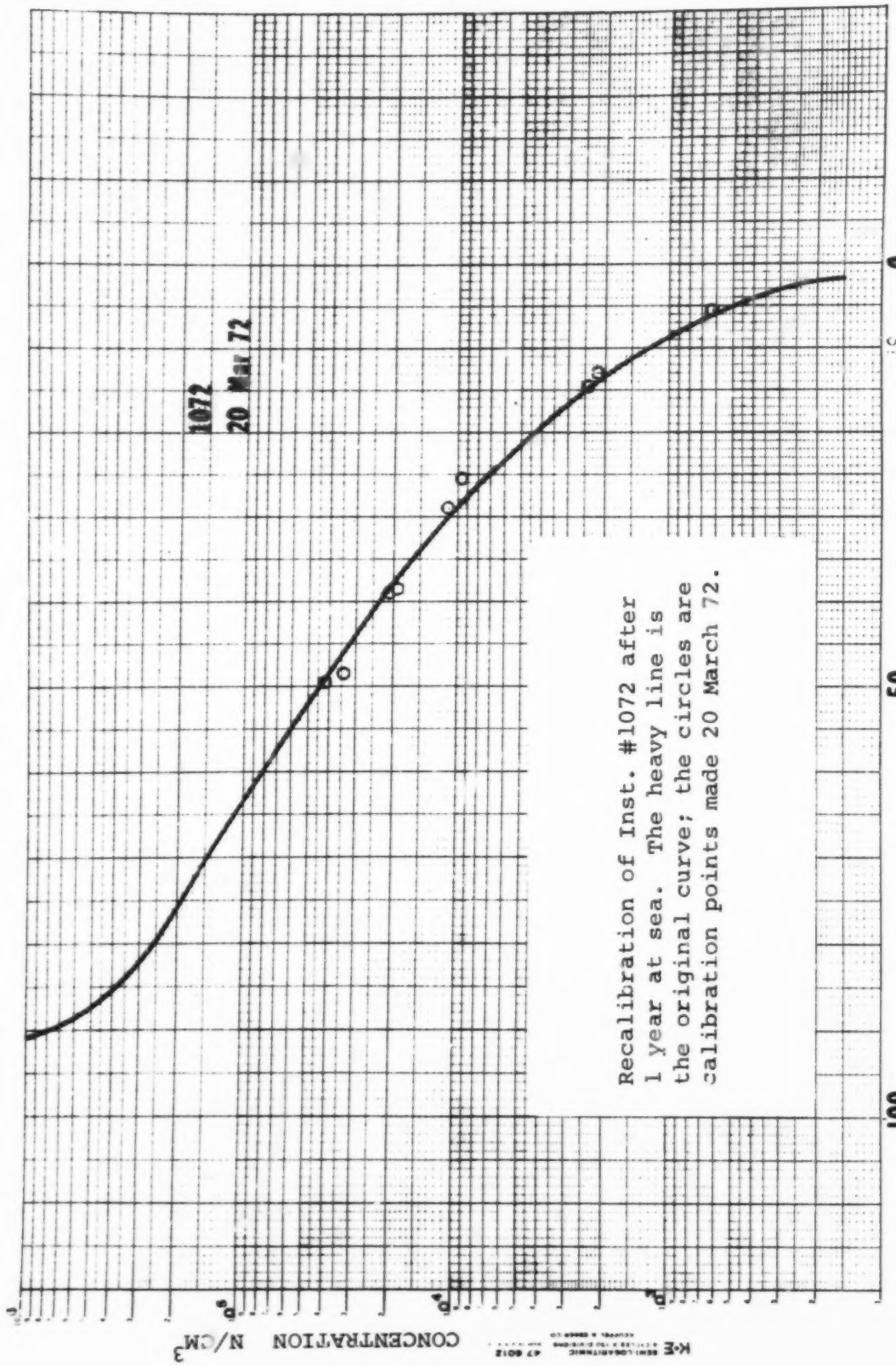
QUALITY OF DATA

The instrument used predominantly in this experiment is the Gardner Associates Small Particle Detector, a slightly modified commercial version of the photoelectric Aitken counter developed by Rich (1955). The instrument has great advantages of portability and compactness, requiring little maintenance. An annual recalibration cycle has been used thus far, and appears to be sufficient in conjunction with semi annual battery changes.

The questions of accuracy and repeatability always arise when such an instrument is operated for long periods under adverse conditions. Because of the dearth of standardized aerosol instruments "accuracy" and "repeatability" must be stated with respect to the Pollak Model 1957 photoelectric counter, using the Pollak and Metnieks (1960) calibration. The original and a repeat calibration of Instrument Serial 1072 are shown as Figure II. The original calibration, performed by the manufacturer in March 1971 is shown as a plotted line; points obtained in re-calibration, following recovery of the instrument from the 'Austral Patriot' in March 1972 are shown as circles. The repeatability of the calibration is within 1 or 2 scale divisions, which is close to the initial ability of the instrument. It would appear then, that the practical field accuracy of the instrument is of this order.

The sampling accuracy is limited by entrainment of some ship air around the superstructure when making observations. When

FIGURE II


1072
20 Mar 72
Recalibration of Inst. #1072 after 1 year at sea. The heavy line is the original curve; the circles are calibration points made 20 March 72.
0
50
100
SCALE READING
CONCENTRATION N/CM3

some relative winds are encountered, it is quite difficult to obtain air free from contamination. Usually uncontaminated air can be obtained by extending a tube slightly upwind of the superstructure, from the upwind wing of the brdige.

Reviewing the data obtained thus far it appears that oceanographic ships often find lower concentrations than merchant ships in the same general area. This is probably because many merchant ships follow the same lanes, and slightly contaminated air might be encountered on occasions when other ships are in proximity although not in sight. While these contaminated readings are no doubt present in the data, the long term averages are probably raised only slightly by inclusion of these observations.

The overall quality of the data is good. Occasionally, obviously contaminated readings are obtained, but the frequency with which data are obtained from specific plots of ocean allows one to reject such spurious values by comparison with the preceeding and following observation, and the values obtained in previous crossings.

RESULTS OF EXPERIMENTS - PRESENTATION OF DATA

OBSERVATIONS AT FIXED STATIONS

Observations made on Shemya Island, in the Aleutian Chain, were discussed in the first annual report. Through the kind accomodation of Farrell Lines, an aerosol instrument was supplied to Thomas Christian, weather observer and radio operator at Pitcairn Island (25S x 130W) in the South Pacific. Observations were begun on 1 January 1972 and are continuing.

The observations are made on the upwind shore of the sparsely inhabited island, or atop a hill at the island's radio station. The readings are occasionally communicated to the investigators by radio; written reports are sent via ship to the Canal Zone, and then mailed to Schenectady, on the chance schedule of ships stopping at the Islands. At the time of this writing, data for January - February of 1972 has been received.

The concentrations observed are relatively low (i.e., 85% less than 500 n/cm^3) as would be expected, with many readings below the threshold of the photoelectric aerosol detector. A recent radio communication however, reported concentrations as high as 1000 n/cm^3 in conjunction with heavy seas and breaking waves. A frequency distribution of the concentrations observed during Jan. - Feb. 1972 is plotted as Figure III, yielding a good approximation of a straight line on log - probability paper. The median value is 335 n/cm^3, with 16% of observations less than

FREQUENCY DISTRIBUTION OF AEROSOL CONCENTRATIONS

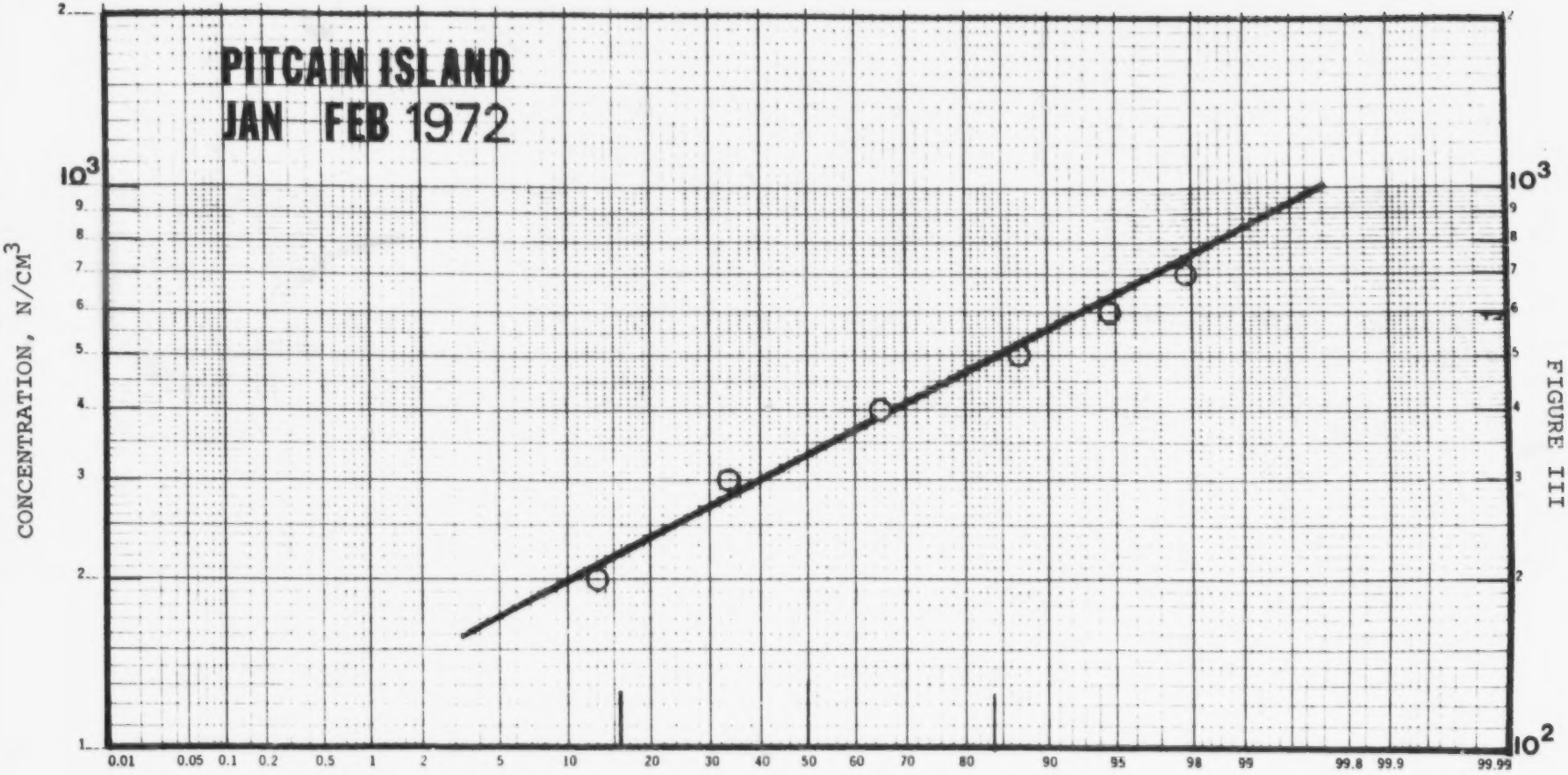

PITCAIN ISLAND
JAN FEB 1972
CONCENTRATION, N/CM3
PER CENT OBSERVATIONS LESS THAN CONCENTRATION
0.01 0.05 0.1 0.2 0.5 1 2 5 10 20 30 40 50 60 70 80 90 95 98 99 99.8 99.9 99.99

FIGURE III

220 n/cm^3. This is in relatively good agreement with a median 260 n/cm^3 reported by an oceanographic ship (F. Guenthner, private communication, 1968) operating in the tropical mid Pacific.

There is not yet sufficient data from Pitcairn to evaluate changes in aerosol concentration relating to seasonal or meteorological variables. This is a prime location for a meteorological observatory, aerosol observations will continue, and solar observations will be added upon arrival of additional instrumentation.

OCEAN STATION

Through the cooperation of Mr. James Dew, NOAA North Atlantic Weather Project, data was obtained at Ocean Station "Echo" during November and December 1971. This station was selected because it is located (35N x 48W) directly east of the continental United States, and slightly south of the area of most frequent ship reports.

Some additional difficulties are encountered when aerosol observations are attempted from ocean station ships, as they are often dead in the water, on station, at the time of observation, with no additional wind over the deck due to the ships' speed through the water. This can cause contamination of readings by ship air as a frequency analysis of all readings obtained at Echo during November - December shows:

CONCENTRATION RANGE - N/CM^3

	300	600	900	1200	1500	2000	3000	4000	
<300	600	900	1200	1500	2000	3000	4000	5000	>5000
18%	27%	14%	4%	3%	0%	5%	1%	3%	13%

The readings below 900 n/cm^3 are probably uncontaminated maritime air; the readings above 5000 n/cm^3 undoubtedly reflect serious

ship air contamination. The sparcity of values between 1500 - 5000 n/cm^3 indicate that periods when only a small amount of ship air is entrained are relatively infrequent; moreover, this partial contamination results in scattered readings which are a signal of shipboard contamination to the observer. When observations of over 1500 n/cm^3 are eliminated from the data, the frequency distribution plotted in Figure IV results. This is a reasonable approximation of a log normal distribution, with a median concentration of 460 n/cm^3. This median, and the average value of 562 n/cm^3 are a good approximation of the values obtained by moving ships in this region during the last several years, as shown in subsequent sections.

When this criterion of rejecting all values above 1500 n/cm^3 as contaminated is applied to a grouping of the data as a function of time of day, a pattern emerges:

Time, Local Standard	0248	0848	1448	2048
G M T	0600	1200	1800	2400
Average Concentration	520 n/cm^3	525 n/cm^3	582 n/cm^3	626 n/cm^3

The concentrations observed at night, and in early morning are lower than those observed during afternoon and early evening. While this apparent diurnal variation is of small amplitude when compared to that which occurs over land, the time of the peak is in agreement with that obtained by the "Empire State" (Hogan et al, 1971, 1967) in nearby waters during summer. This diurnal change may be indicative of solar conversion of certain vapors to particles, or of mixing of particles or ozone from higher altitudes with surface air.

FREQUENCY DISTRIBUTION OF AEROSOL OBSERVATIONS

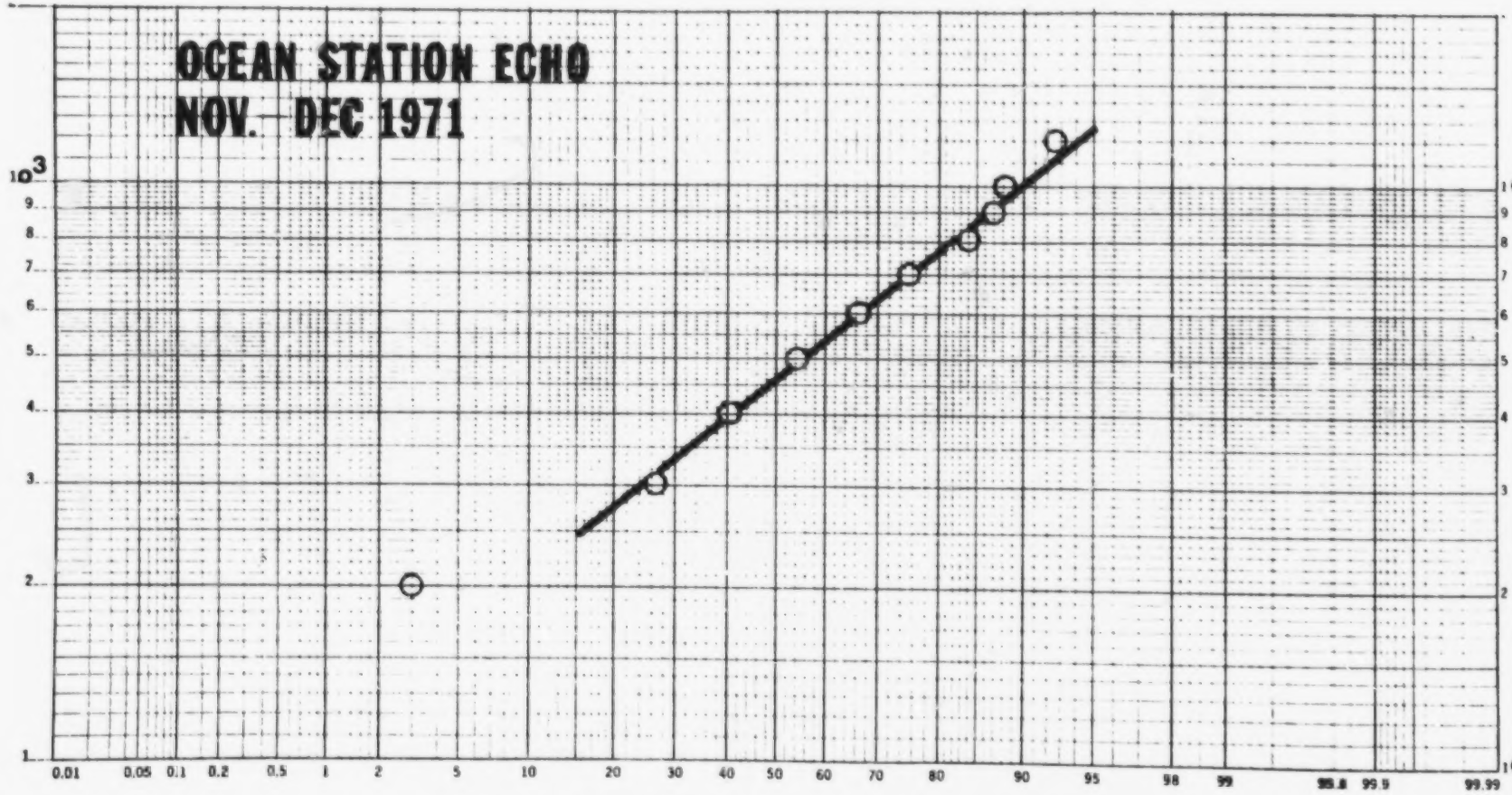


FIGURE IV

Grouping the data with respect to wind direction at the time of observation yields a "rose" with its longest significant vectors pointing west and north:

Wind Direction Degrees	030	090	150	210	270	330
Number Observations	9	4	14	10	11	20
Average Value CN/CM^3	730	400	481	540	699	543

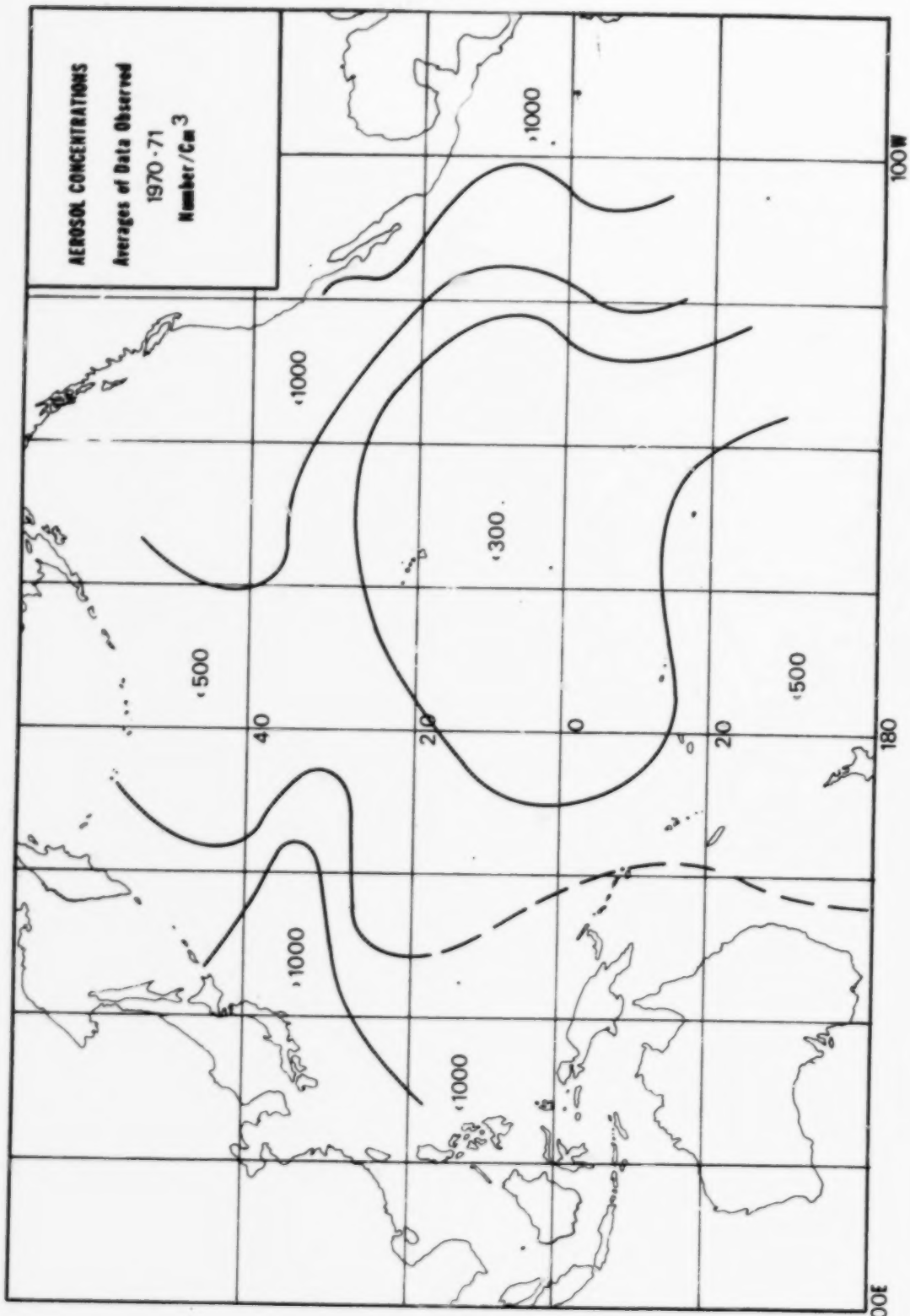
An analysis of observed concentrations as a function of wind velocity did not yield the smooth curve found North of $40^{\circ}N$ during June; however, wind velocities less than 6 M/sec., where this trend was observed, were infrequent at Echo during this period.

AEROSOL CONCENTRATIONS OVER THE PACIFIC

Aerosol observations over the Pacific are primarily from the Panama Canal - Japan Great Circle route, the Panama Canal - Australia route, and the Los Angeles - Honolulu - Hong Kong - Yokohama - San Francisco run, which comprise a fair network, but leaves great areas unexamined. These routes have been supplemented by "one time" passages of the "Robert D. Conrad," of Lamont, in 1968, and the "Thomas Washington" of Scripps in 1972, and the recent Pitcairn Island data.

The numerical average of all observations made in each $4^{o} \times 4^{o}$ block from which data is available has been computed, and entered as a function of position, on a mercator projection. Isopleths enclosing areas averaging less than 300 n/cm^3, less than 500 n/cm^3, less than 1000 n/cm^3, and more than 1000 n/cm^3 were then faired, to produce Figure V. Because of the seasonal variation of winds in the vicinity of Japan, extreme variability was found in this area. Concentrations as low as 280 n/cm^3 were measured a few miles offshore with the wind from the sea, much in the same way low concentrations are often found at the Atlantic end of the English Channel. With wind from the land, concentrations in the thousands are found several hundred miles East of Japan. This extreme variability is the reason for grouping the data in a catchall >1000 class, rather than fairing higher concentration isopleths, as is done with the more frequent Atlantic data.

FIGURE V


AEROSOL CONCENTRATIONS
Averages of Data Observed
1970-71
Number/Cm3
1000
1000
300
500
500
1000
1000
40
20
0
20
180
100W
100E

The sparcity of data available prevents fairing of finer structure to the isopleths than that shown in the figure, or isolating areas of extreme low concentration as shown by Shiratori in his analysis of Carnegie data. The plot is very preliminary, and will be improved with the addition of the data now being observed in the area.

Examination of the figure shows a great area of low concentration dominating the tropical Pacific. This is surrounded by narrow belts of <500 and <1000 n/cm^3, the only areas of high (i.e., >1000 n/cm^3) concentration being adjacent to the land masses. Moderately low concentrations are found off the California coast, but higher concentrations seem to extend further a-sea west of South America. These may be seasonal results of local circulation, and may change, as a result of more data input. The preliminary data does show the existence of a great reservoir of low particle concentration throughout the Pacific basin.

HAWAII EXPERIMENT FEBRUARY 1972

One of the investigators was privileged to accompany a Naval Research Laboratory research flight in Hawaii in February. Aerosol experiments were conducted in parallel with Cloud Condensation Nucleus experiments of Hoppel and Wojcieskowski of N.R.L., along a transpacific flight path from NAS Point Mugu, California to Hickam Field, Hawaii, over the volcanic areas of Hawaii, and over the open sea 20 miles north of the island of Oahu.

Concentrations measured on the crossing ranged from 150 to 300 n/cm^3 at 6000'msl. Low (<300 n/cm^3) concentrations were obtained at that altitude, within sight of land, in agreement with mountaintop measurements in California during that season in previous years. The lowest measurements were near the threshold of the special (Hogan and Gardner 1967) photoelectric nucleus counter used, and may have been somewhat lower.

A very interesting series of concentration versus altitude profiles were obtained 20 NM north of Oahu on 6-7 February. Near surface observations were comparable to those obtained by Blanchard at the shore, in previous experiments, and by the investigator at the same site following the airborne experiments. Concentrations measured at higher altitudes were considerably higher, and varied by a factor of three at 15000 ft. on 7 February:

ALTITUDE, MSL	CONCENTRATION			
	TIME	6 FEB. 1972	TIME	7 FEB. 1972
15000'	1506	300 n/cm^3	1555	1080 n/cm^3
13000'			1618	310
11000'			1621	350
9000'	1411	200-440 n/cm^3	1630	410
8000'			1639	880
7000'	1340	200-220 n/cm^3	1644	390
APPROXIMATE LEVEL OF TRADE INVERSION				
5000'			1651	170
3000'			1656	170
2000'	1300	150-180 n/cm^3	1703	120
1000'			1706	180
500'	1251	150-180 n/cm^3	1711	120

It appears that removal mechanisms are quite effective in removing near surface aerosols over the Pacific but are less effective above the inversion. Contamination cannot be completely ruled out in this experiment, as it was conducted in a flight training area, but no other aircraft were sighted during the experiment. Surface observations at Hawaii detect the presence of an aureole around the sun, when surface aerosol concentrations are low; this supports the existence of greater aerosol concentrations at higher altitudes. Some further aircraft experiments, over the open sea, are necessary to determine if higher concentration is permanent, and if it is the result of transport from the continents or up from the surface, or, if it is the result of atmospheric chemical reactions in this bright sunlight.

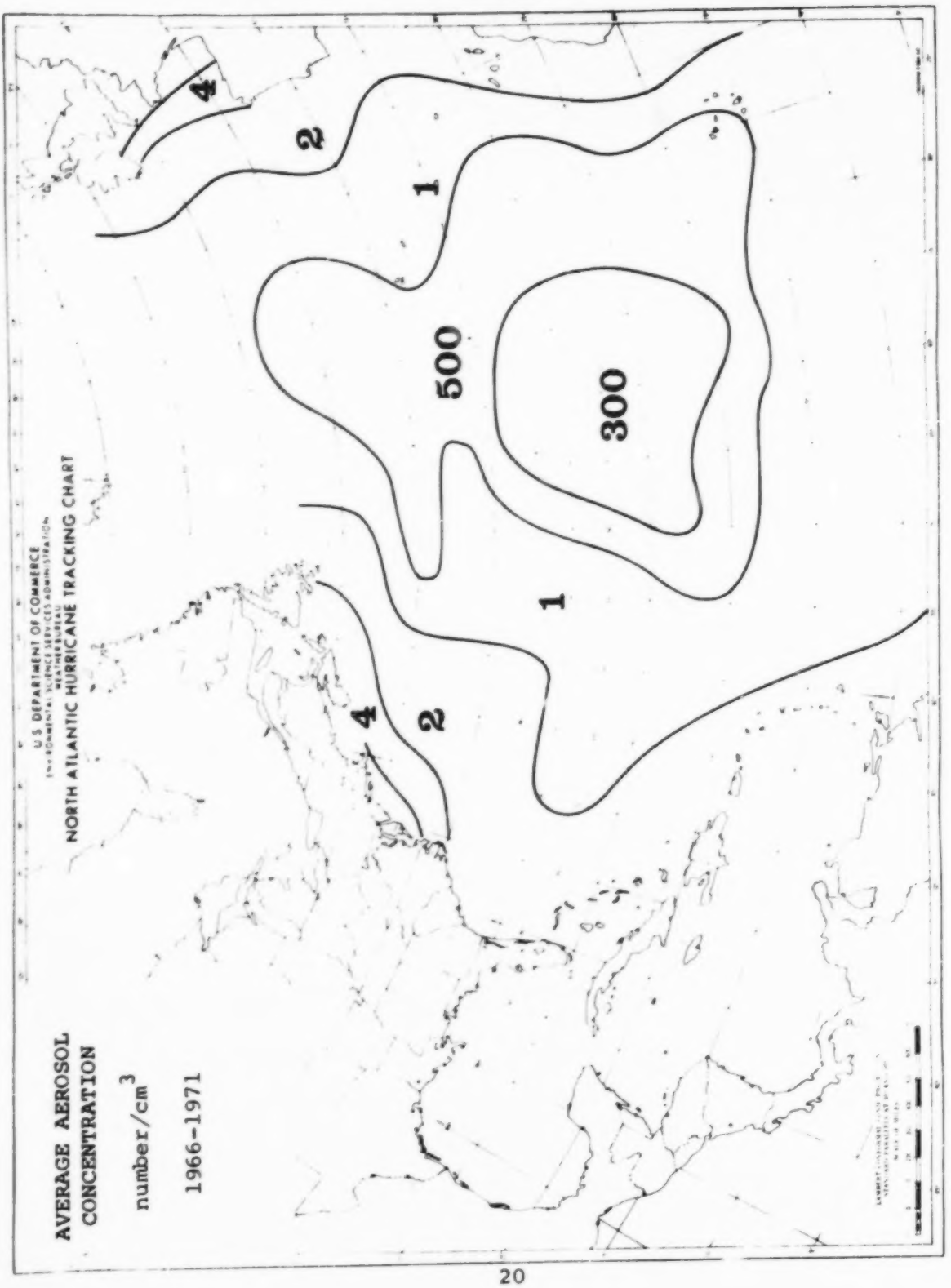
AEROSOL OBSERVATIONS OVER THE ATLANTIC

The most frequent data thus far available is from the North Atlantic Ocean, south of 50N latitude. This area is densely covered by the "Empire States" 40°N crossings, trade routes from New York to Northern Europe, Africa and South America, and supplemental data from Oceanographic ships and ocean station. "Empire State" data began in 1966, and New York - Africa data is available from 1968; all other routes commenced in 1970. The North Atlantic data has been analyzed in several ways, with respect to position and chronology.

A. The distribution of aerosol concentrations over the North Atlantic.

A preliminary map showing the average aerosol concentrations was prepared for the first annual report. Additional data obtained since that time has allowed computation of new average aerosol concentrations for 4° Lat. x 4° Long. blocks, and fairing with greater certainty, isoplethes enclosing average aerosol concentrations of less than 4000, 2000, 1000, 500 and 300 particles per cubic centimeter. The isoplethes of average aerosol concentration are shown in Figure VI plotted on a lambert conformal conic. Examination of the figure shows that, on the average, continental type aerosol overpowers the maritime regime for several hundred miles in the Bermuda region, and along a narrower belt west of the coast of Europe and Africa. On the average, a small region

FIGURE VI


AVERAGE AEROSOL CONCENTRATION
number/cm3
1966-1971
U.S. DEPARTMENT OF COMMERCE
NORTH ATLANTIC HURRICANE TRACKING CHART
4
2
1
500
300
1
2
4

of relatively low (<500 n/cm^3) aerosol concentration separates the regions of relatively high (for oceanic concentrations 500-1000 n/cm^3) found around the Azores and east of Bermuda. During some periods, however, a high concentration band crosses the Atlantic along this Bermuda - Azores high pressure belt, as witnessed during 1971 crossings of the "Empire State."

The concentrations plotted in Figure VI are averages; two additional maps, showing concentration extremes are attached as an appendix. These extremes show that aerosol concentrations at or below the threshold of the photoelectric counter have been observed over most of the Atlantic during the past two years, including within the English Channel and near the coast of North America. High (relatively) concentrations can also be found from time to time in most areas; however, no concentration greater than 1000 n/cm^3 has been found in mid Atlantic since 1966.

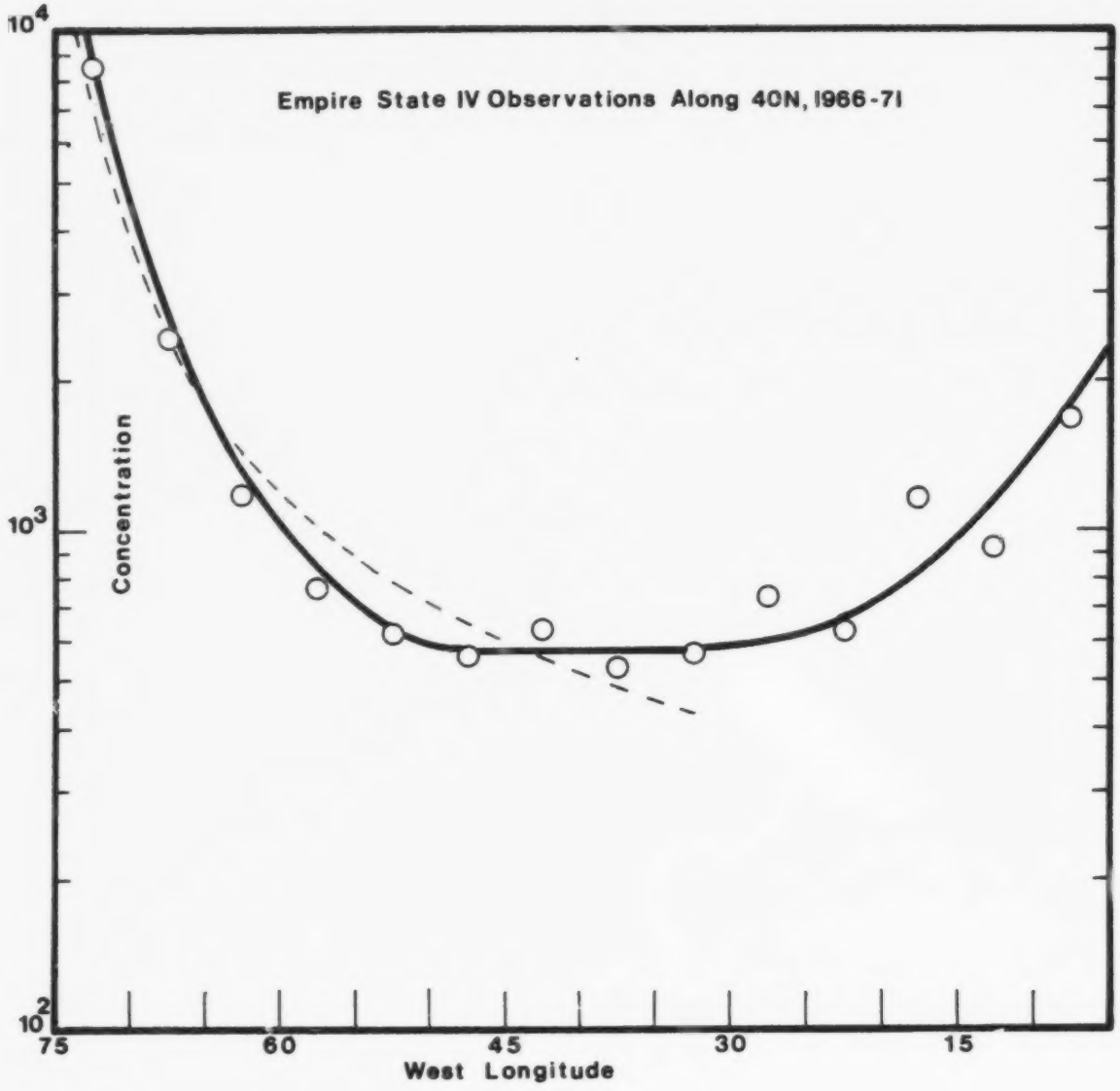
An area of low (<300 n/cm^3) aerosol concentration was found along the New York - English Channel Great Circle route, during the first few observations made during this program and reported in the first annual report. A much higher value was found with the addition of the second years' data. This may be a seasonal effect, as year two's data reflect many spring, summer and fall observations, or it may be related to more complicated circulation patterns, as observations along 40°N were higher on the average, in June and July of 1971 than in previous years.

An additional analysis was performed on the 1966-1971 "Empire State" data, observed from 35-42N. This data is quite homogenous, all obtained during summer months, along quite similar routes, and is the longest run of record available. The data was broken down into 5° longitude blocks, and the average of all data obtained in each block calculated, and plotted in Figure VII as a function of longitude.

The average concentration falls off rapidly with distance from shore in the western Atlantic, remains relatively low and constant east of 55°, and rises gradually east of the Azores. A previous analysis of one traverse of fair weather data along this route (Hogan 1970) showed that the concentration could be expressed as a hyperbola of form X (distance) · Z (concentration) = K (constant). As a great majority of the 1966-71 data was observed during fair weather, a hyperbolic fit was attempted for the average values. The constant K was calculated for each average concentration value (i.e., in the 70-75° block, the average point is 150 miles offshore, in the 65-70° block 450 miles, etc.) and the values of K determined to 30°W, where Azores influence terminates the analysis, and an average value for K computed. A hyperbolic decay was then calculated, as shown in Figure VII by the dotted line, using this average K, and the mean distance in each block. The fit of this calculated curve to the data is reasonable, if not exact.

If one assumed the North Eastern United States to act as a bounded line source of aerosol, which then was two dimensionally

FIGURE VII


Empire State IV Observations Along 40N, 1966-71
Concentration
10^4
10^3
10^2
75
60
45
30
15
West Longitude

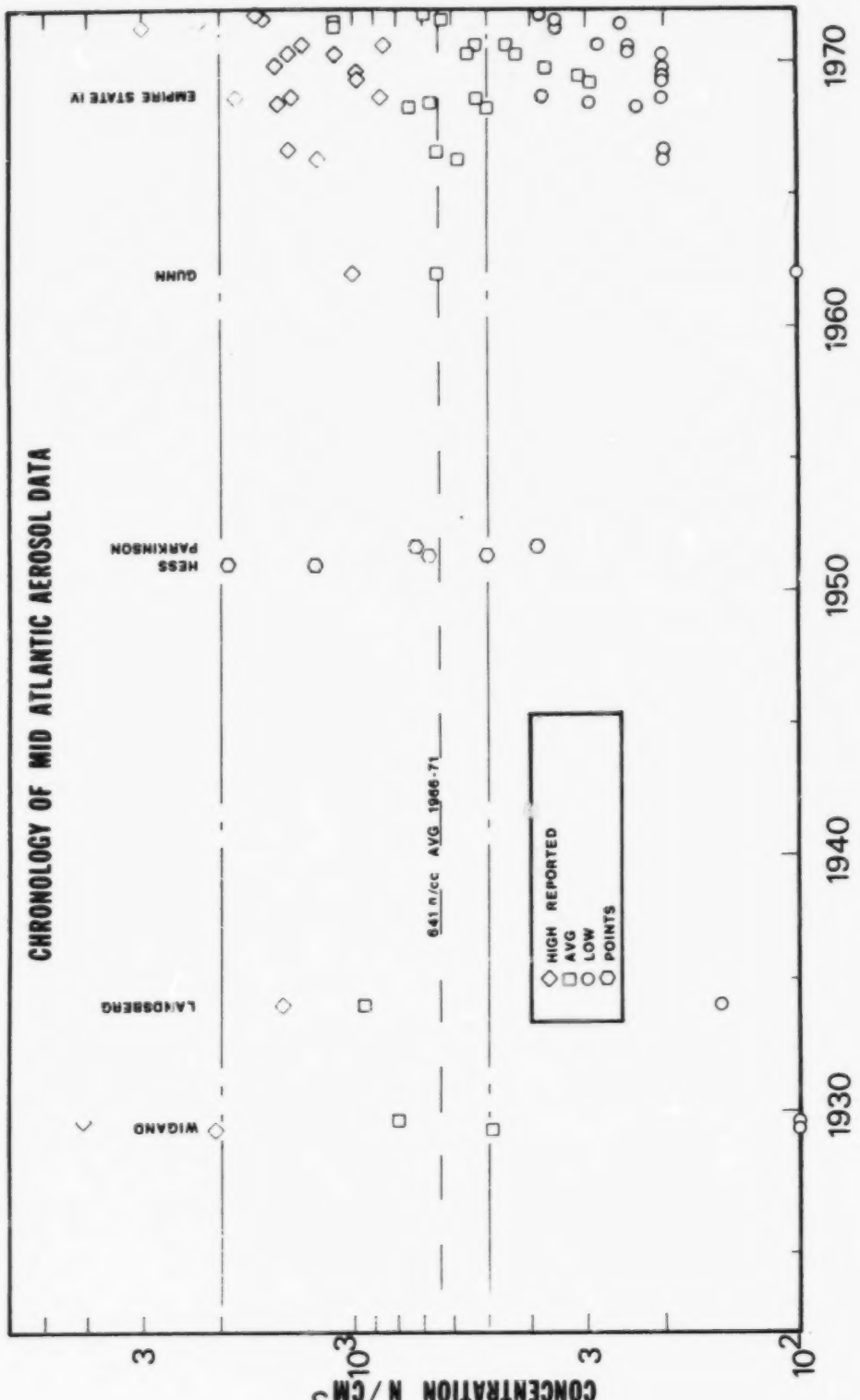
diluted under an inversion or other "lid," with the dilution volume increasing linearly with distance, an XZ=K hyperbolic decay of aerosol concentration such as this would then be expected. The departure of observed values below that predicted between 60° and 45° might be indicative of some coagulation reducing the overall concentration; the relatively constant levels found east of 45° might indicate that the major source was bounded by minor line sources, and further dilution could not occur. The relatively constant level observed east of 45° might also be indicative of a constant local production resulting in such a background in the absence of removal mechanisms such as clouds or storms. This relatively simple model seems to be quite useful in predicting offshore concentrations in the absence of storms or other major cleansing processes.

B. Chronology of Atlantic aerosol data.

A chronology of "Empire State" observations made in the mid-ocean region bounded by 35-42°N latitude and 30-60°W longitude has been prepared. The average value obtained from all data observed in this block during 17 individual crossings is 641 n/cm^3. Average values within $\pm$10% of this long term average were obtained on five (5) individual crossings; higher values were obtained on three (3), and lower on nine (9) crossings. No great or permanent trend appears in the data; the low values found in 1969 were followed by average values in 1970, and the extremely high values observed in June and early July of 1971 were immediately followed by average values in late July and August of the same year.

The values obtained during these recent experiments have been plotted chronologically with the historic data of the "Carnegie" (Shiratori, 1934), Wigand 1929, Landsberg 1938, Hess 1951, Parkinson 1952, and Gunn 1964. The average, high, and low value, or points reported by these authors are plotted in Figure VIII, with the 641 n/cm^3 average of the "Empire State" 1966-71, and the maximum and minimum (500 and 2000 n/cm^3) of Shiratori's calculations delineated. Where possible, only data obtained between 30-60°W longitude and 35-42°N latitude was included to permit direct comparison with the "Empire State" data; however, in some cases it was not possible to eliminate all near shore data. The average value of 900 n/cm^3 quoted by Landsberg, for example, probably contains some nearer shore readings. The values have a tendency to group around the recent average of 641 n/cm^3; the differences are probably well within the comparability of the various instruments used, in the concentration range observed.

FIGURE VIII


CHRONOLOGY OF MID ATLANTIC AEROSOL DATA
EMPIRE STATE IV
GUNN
HESS PARKINSON
LAKOSBERG
WIGAND
641 n/cc AVG 1966-71
HIGH REPORTED
AVG
LOW
POINTS
1970
1960
1950
1940
1930
CONCENTRATION N/CM3
3
10^3
3
10^2

DISCUSSION OF DATA

A review of the maps showing average aerosol concentrations (Figs. V and VII) and the appendix Figs. II and III showing the extremes of aerosol concentration, show that, in all instances, lowest concentrations of aerosol are found in mid ocean, and the highest near shore. A relatively smooth gradient exists from areas of high to low concentration on the average, and a tongue of continental aerosol extends far offshore in the Bermuda region in the Atlantic and east of Japan in the Pacific. Mid ocean level aerosol concentrations are found very close to shore during periods of cloudiness or precipitation, and when the air mass has had a long overwater trajectory. High concentrations are found far at sea in times of clear, stable weather. These high concentrations decrease with increasing distance from land, and as shown in Figure VIII follow essentially a hyperbolic decay, to mid ocean, east of the densely populated areas of the United States.

It is indeed evident that the continents are the major source of the number concentration of particles found in the vacinity of the continents, and that these particles can be carried far to sea in fair weather. It is also evident that, on the average, these excess particles are removed by mid ocean by some mechanism.

It was stated during the first annual report, that aerosol concentrations were as variable as any other meteorological parameter; additionally, data obtained as part of that research indicated that the cloud forming mechanism was a major sink for small aerosol particles. The recent data accentuates that idea, as extreme variations in aerosol concentrations occurred over the north Atlantic during this period. This variation is very great along $40^{o}N$, and is also seen along the more northerly routes.

The data obtained along $40^{o}N$ is very homogeneous; the same type instrument has been used, on the same ship, along nearly identical routes, during the months of June, July and August for six years. Although the individual observers have changed in alternate years, they are all students in the SUMC meteorology program, under the guidance of that faculty.

The variability obtained along this route is extreme; the average value for a crossing through the "uncontaminated" band between 30-60W has ranged from 293 n/cm^3 in June of 1969 to 1115 n/cm^3 in June of 1971. The June 1969 low as followed by a higher average of 368 n/cm^3 in August, and June 1971 high was followed by an almost exactly average 630 n/cm^3 on the next crossing. Comparing these crossings with historical data from the literature, shows the low averages of June 68, June, July, August 69, June 70 and August 70 to be quite similar to the data obtained by Wigand on the westbound leg of his 1929 expedition. The high values of June - July 1971 are much like those obtained

in 1951 by V. F. Hess. The average obtained by Gunn in 1962 is 650 n/cm^3, almost exactly the long term average of 641 n/cm^3 obtained by the "Empire State" in the years 1966-71. The other historic data of Wigand, Landsberg, Hess and Parkinson all parallel one or more crossings of the "Empire State."

This historic data may not be directly comparable to the "Empire State" data, as a photoelectric nucleus counter was used for all "Empire State" programs, and "absolute" counters of the Scholz and Aitken types were used by previous investigators. Also, the "Empire State" data has been obtained systematically at scheduled times in a well defined area. The historic data was often obtained by the observer from his cruise ship, while on his way to or from scientific meetings, or as an adjunct to atmospheric electricity measurements, and not taken on regular schedule. The routes followed and the season of crossing was also more variable in the case of the prior investigators but where possible, data obtained north of 45°, and near shore has been eliminated before the points reported in this paper were computed. While it is not possible to directly compare this data as a chronologoly, it is certainly evident that aerosol concentrations over the north Atlantic are extremely variable.

The previous discussion has shown that when high numbers of particles are found at sea, they are probably of continental origin. The instrumentation used does not allow one to determine the nature of the individual particles; they may be the result of fossil fuel

combustion, natural combustion such as forest fires, or the result of chemical reactions in the atmosphere involving both natural and man made sulfur dioxide, hydrocarbons, or other vapors (Went 1960, Lodge and Mohnen 1969, Schaefer 1970). While no chronology of natural hydrocarbon production or forest fire fuel combustion exists, one can extract a chronology of fossil fuel consumption from the Pocket Data Book of the United States.

YEAR	TOTAL FOSSIL FUEL CONSUMPTION, MILLIONS OF TONS
1929	800
1934	596
1948	1051
1950	1002
1951	1048
1960	1181
1965	1399
1967	1535
1968	1584
1969	1643

While monthly or seasonal trends cannot be extracted from these numbers, they do illustrate that there was a general decrease in fossil consumption during the depression of the 30's, and that annual fuel consumption was relatively constant from 1965-69. Great variability in Atlantic aerosol concentration was found during the period of relatively constant fossil fuel consumption by the "Empire State," and Landsberg's high average of 950 n/cm^3 was found during the period of lowest fuel consumption.

It might be argued that great changes in natural production of small aerosols and convertible vapors may have occurred during these periods, causing this great variability during times of

relatively constant fossil fuel burning. The tongues of highest concentration shown in the figures extend from the areas of high fuel consumption; but this is not a unique argument, as a northwest flow is rather common at that latitude.

It would then appear that the meteorology is the dominant mechanism in establishing aerosol concentrations at the surface. Variability in low level winds, and changing trajectories would be a direct and simple explanation; creation of additional particles by ozone reactions in subsident air might also be an explanation, were sufficient data available. Recent satellite data (Global Atlas of Relative Cloud Cover 1967-70, USDC-USAF, Washington 1971) shows that a steep gradient in average cloud cover exists along 40°N during the months of June, July and August. It may very well be that the frequent changes in cloud cover, which must accompany this steep gradient, may be a dominant factor in aerosol removal and produce the frequent short term variations observed.

It would seem that a "model" of the atmospheric surface aerosol could be stated: Large numbers of small aerosol particles are generated by fossil fuel combustion and natural fires over the continents; simultaneously certain gases are liberated by these combustion processes. Additionally, similar gases are liberated by natural plant processes, and anthropogenic processes, which will, eventually be converted to particles by reaction with sunlight, ozone, or other gases. The mixture of atmospheric gases and

particles is carried out to sea by the prevailing winds, where it mixes with less continentally influenced air, and the concentrations of both gases and particles are diluted. In high pressure systems, in the absence of low cloudiness, gases are converted to particles by sunlight, reaction with ozone or other oxidants in subsiding air, and the number concentration is reduced only by dilution or coagulation.

When the initial aerosol, and that formed later by reactions becomes involved in a forming cloud, a small number of "active" (or large) particles immediately become the nucleus of cloud droplets. During the clouds formative period, while drops are growing and the water vapor flux is toward the drop, large numbers of small particles collide with the drop through Brownian diffusion. These small particles are then trapped within, or, in the surface film of the drop. If the drop continues to grow, and falls as part of the frequent precipitating fog or drizzle frequently found at sea, all of these particles and their gaseous precursors will be removed. If the drop evaporates before precipitating, the small particles will all be firmly agglomerated to the remaining single nucleus, leaving one particle in the volume formerly occupied by many.

The maps and profiles presented with this paper verify the continental source of large numbers of aerosols. Experiments aboard the "Empire State" and "F. S. Meteor" have shown that

lowest aerosol concentrations are found in precipitating fog or drizzle, and that aerosol concentrations 200-300 n/cm^3 (i.e., the same order as the number of drops in a cloud) are found to the seaward side of a frontal system. The weakest point in this theory, experimentally, is the formation of additional aerosols from convertible gases. The only evidence for this occurrence in nature is the small diurnal variation found by the "Empire State" and Ocean Station Echo.

CONCLUSIONS AND RECOMMENDATIONS

1) The continents are the major source of large number concentrations of aerosols. While great quantities of aerosol are generated by bubbles and breaking waves, the influence of continental aerosol at sea is unmistakable.

2) The number concentration of aerosol particles is generally reduced to background by mid Atlantic, although occasions do occur, concurrent with the Bermuda-Azores high build up, when concentrations approaching 1000 n/cm^3 are found to bridge the Atlantic.

3) A hyperbolic approximation is a quite accurate tool for prediction of clear weather aerosol concentrations east of the United States. The average summer value of aerosol concentration at 40^{o}, from 75^{o}W to 40^{o}W is given by Z (concentration) $\cdot$X (distance) $= K = 1.07 \times 10^{6}$.

4) Cloudiness, especially at low levels, and fog seem to be an important sink for surface concentrations at low levels.

5) Aerosol concentrations, at sea, are extremely variable. The variation about the mean measured during extensive experiments of the last five years, is similar to the variation measured in the last forty three years. A secular variation in aerosol concentrations may very well exist, but could only be measured by a

systematic observation program, using frequently standardized aerosol detectors, of high resolution and accuracy.

6) Aerosol data obtained by cooperating observers aboard merchant ships is of good quality, although it may be slightly contaminated by frequent passage of many ships through narrow lanes. Aerosol data from cooperating observers on oceanographic ships and ocean station is also of good quality, although contamination may occur when the ship is stationary. A valuable synoptic aerosol network could be established by supplying simple instruments to be used in conjunction with NOAA synoptic observations aboard these ships.

7) The nature of this program is such that additional data is still being received, although the program is officially over. Additional analysis and updating will be performed, and results will be available to EPA Division of Meteorology on request.

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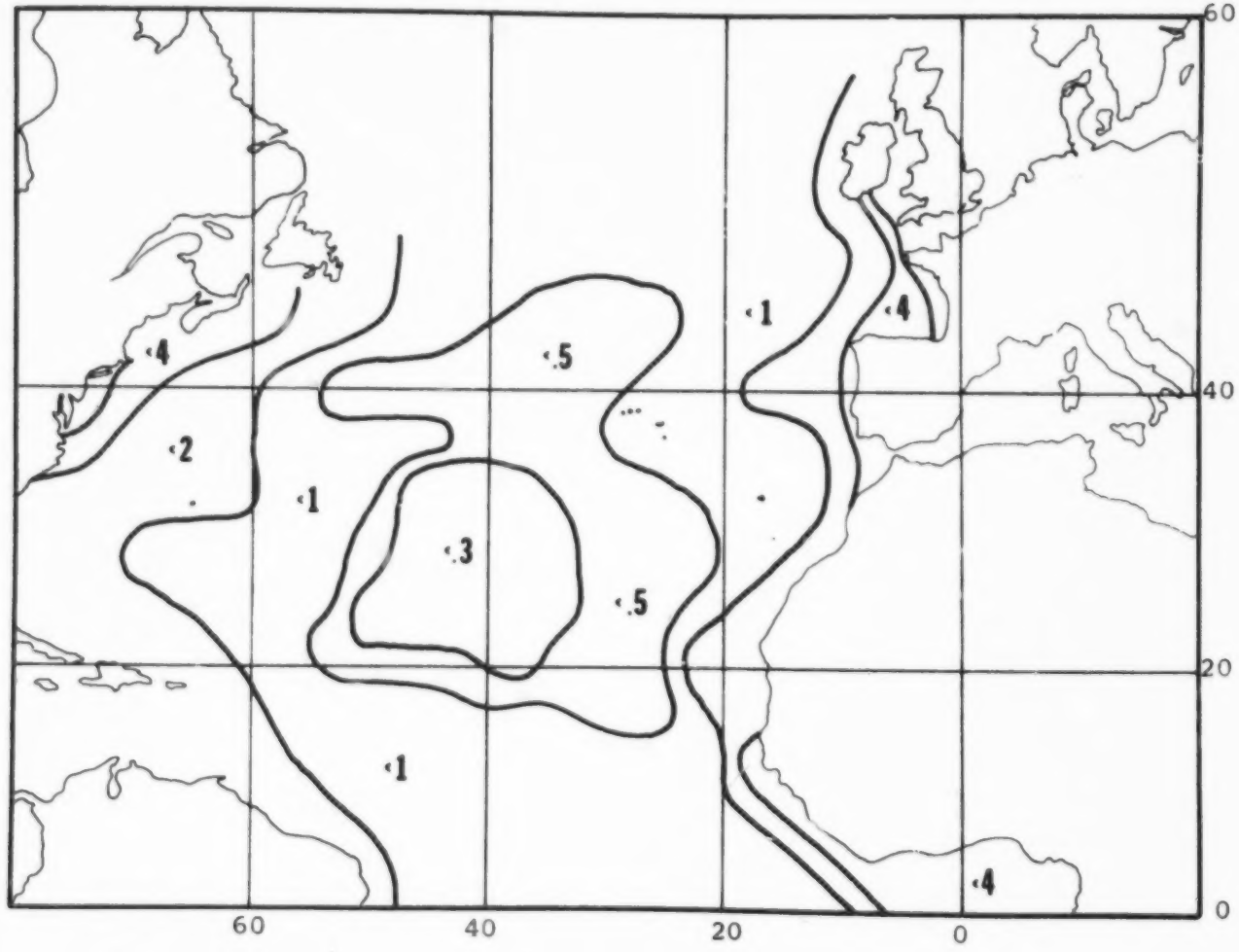
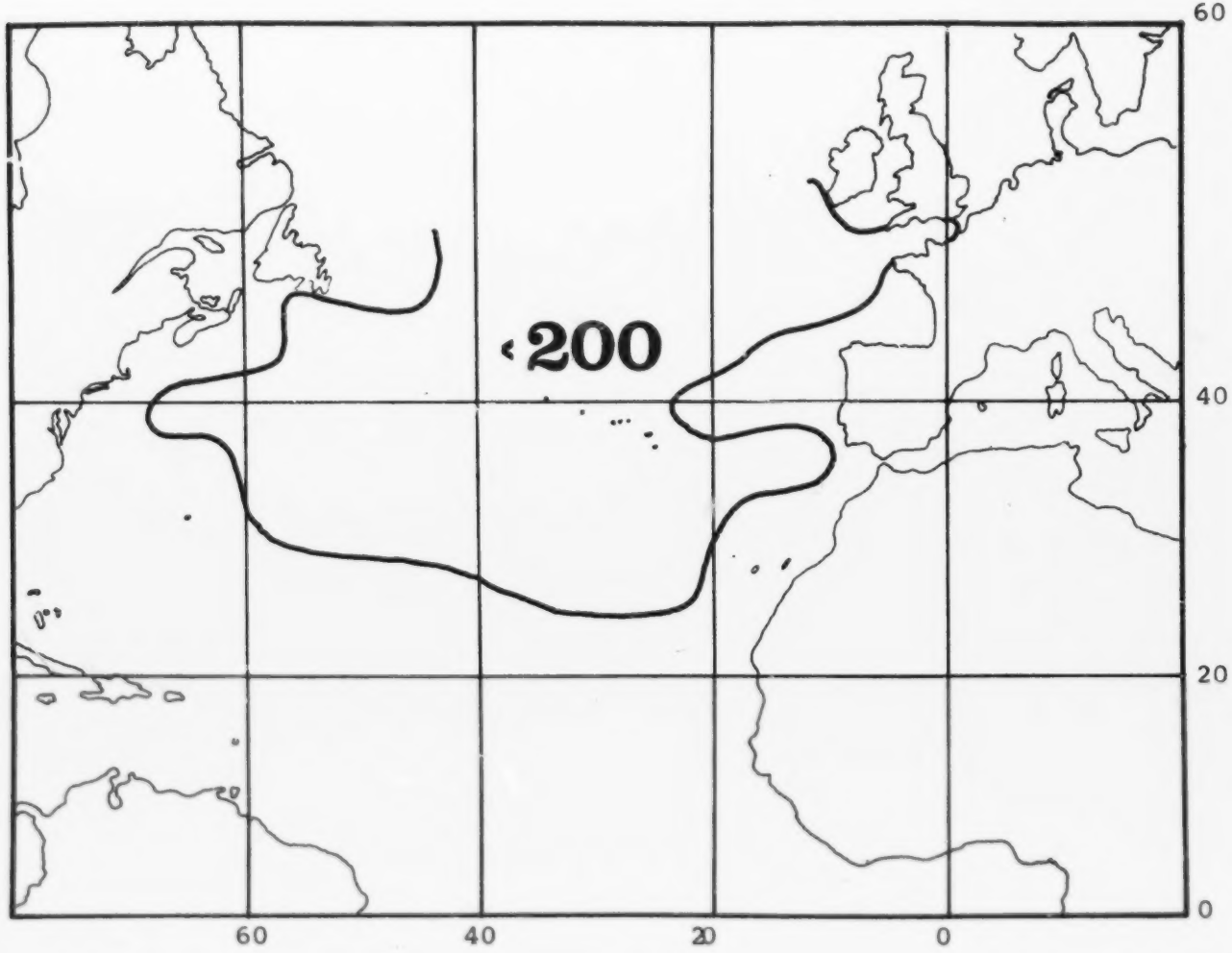
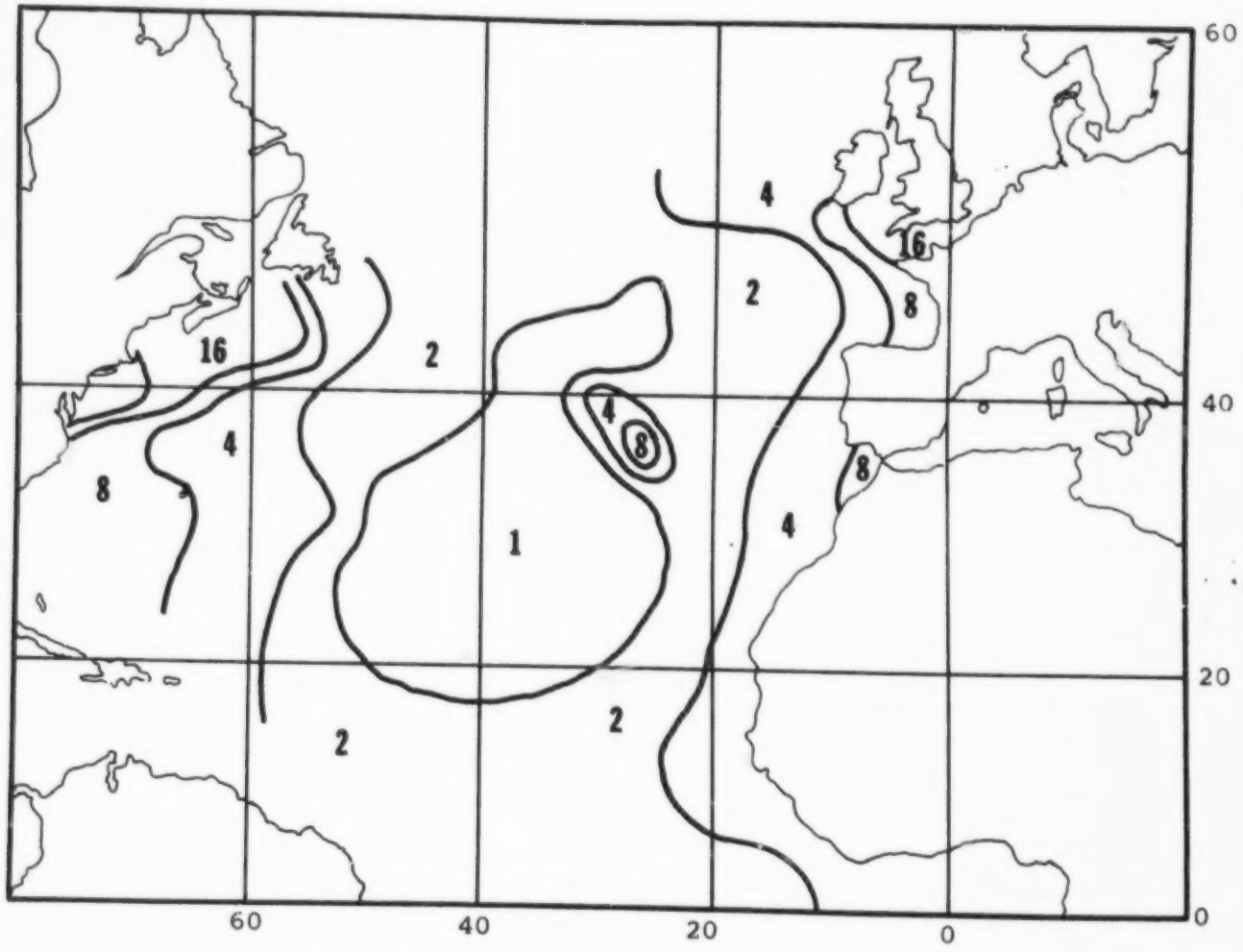


APPENDIX FIG. I. ISOPLETHES OF AVERAGE AEROSOL CONCENTRATION PLOTTED ON MERCATOR PROJECTION



APPENDIX FIG. II. THE HEAVY LINE ENCLOSES THE AREA WHERE AEROSOL CONCENTRATIONS AT OR BELOW THE THRESHOLD OF THE PHOTOELECTRIC COUNTER HAVE BEEN OBSERVED, 1966 - 1971



APPENDIX FIG. III. ISOPLETHES ENCLOSING THE HIGHEST AEROSOL CONCENTRATIONS ENCOUNTERED 1966 - 1971 IN THOUSANDS OF PARTICLES PER CUBIC CENTIMETER

K•E 4 X 4 TO THE INCH 46 0253
7 X 10 INCHES MADE IN U.S.A.
KEUFFEL & ESSER CO.

APPENDIX TABLE I

AVERAGE VALUE, AND NUMBER OF OBSERVATIONS USED TO PREPARE NORTH ATLANTIC MAP.

58	50	42	34	26	18	10

K·E 4 X 4 TO THE INCH 7 X 10 INCHES 46 0253 MADE IN U.S.A. KEUFFEL & ESSER CO.

APPENDIX TABLE II
HIGH AND LOW CONCENTRATIONS OBSERVED OVER ATLANTIC